Uniform deposition of $YBa_2Cu_3O_7$ thin films over an 8 inch diameter area by a 90° off-axis sputtering technique

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(Received 8 February 1996; accepted for publication 16 October 1996)

The uniform deposition of $YBa_2Cu_3O_7$ (YBCO) thin films over an 8-in.-diam. area, using a 3-in.-diam. sputtering target and optimized substrate rotation in a single target 90° off-axis sputtering technique, is reported. Two dimensional maps of the thickness profile of YBCO films deposited on a stationary substrate have been obtained using surface profilometry. These thicknesses were used in a computer simulation to predict which distance of the target from the center of the substrate rotation will produce the maximum area with uniform thickness. The films deposited on substrates mounted on a rotating arm displayed uniform thickness ($<\pm 5\%$ variation) and composition ($<2.3\%$ deviation from the target stoichiometry) and a consistently high transition temperature ($T_c > 87.5°$ K) and critical current density ($J_{c\ 4.2\ \mathrm{K}} > 2\times 10^7$ A/cm^2) over an 8-in.-diam area. © *1996 American Institute of Physics.* [S0003-6951(96)02551-X]

Epitaxial thin films and heterostructures of oxide materials have great potential for novel device applications because these materials exhibit an enormous range of electrical, magnetic, and optical properties. Large area uniform film deposition is required to get reliable device performance over the entire wafer and for efficient processing. Several techniques have been successfully used to grow various oxide thin films on small areas ($\leqslant$3 in. diam), such as pulsed laser deposition,[1] metal organic chemical vapor deposition (MOCVD),[2] cylindrical magnetron sputtering,[3] on-axis dc magnetron sputtering,[4] 90° off-axis sputtering,[5,6] ion beam sputtering,[7] and multi gun off-axis sputtering.[8,9] Recently, Kinder *et al.*[10] have used thermal reactive evaporation from elemental sources, in conjunction with a rotating disc heater which allows intermittent deposition and oxidation in spatially separated zones, to deposit very large area ($\approx$9 in. diam) YBCO films.

90° off-axis magnetron sputtering[6,11] has been especially attractive and widely used for large area deposition of oxide thin films because of its simplicity and reproducibility. It has already been demonstrated that this process can produce thin films with the smooth surfaces required for multilayer growth and device applications.[6] This technique also uses a single composite target instead of multielemental sources, which is a considerable advantage in heterostructure and multilayer deposition. However, the growth rate is relatively slow and acceptable thickness uniformity has been demonstrated only over 3-in.-diam areas.

In this letter, we present the results of very large area deposition of YBCO thin films using a larger sputtering target and optimized substrate rotation in a single target 90° off-axis sputtering technique. The film thickness distribution that is obtained from a sputtering source is primarily dependent upon the shape and size of the erosion zone of the target, which is annular shaped in planar magnetron sputtering. It is thought that increasing the diameter of the erosion ring will cause more lateral spread of the flux. Hence, better thickness uniformity should be obtained in the lateral direction. Furthermore, due to a larger erosion zone area, there should be an increase in the flux emitted from the target provided that the current density is kept constant. As a result, the growth rate should increase.

Figure 1 shows the 90° off-axis sputtering geometry and the various reference axes on the substrate along which the variation in thickness, composition, and superconducting properties were measured. The $X-X'$ axis is the lateral axis along which the distance from the target is fixed, and the $Y-Y'$ axis is the longitudinal axis along which the distance from the target increases. We have mounted both a standard 2-in.-diam. and a larger 3-in.-diam. stoichiometric YBCO target on planar magnetron US′ Gun II sputter sources. All the films were deposited at an operating pressure of 200 mTorr (80% Ar/20% O_2) while maintaining a constant distance h between the substrate and the edge of the target (Fig. 1).

To maintain consistency in the experiments, both targets were sputtered at the same rf power density. The shape and size of the erosion rings of the two targets were measured after presputtering. The mean diameter of the erosion groove of the 3 in. target was found to be 1.7 in. and that of the 2 in. target 1.1 in. As a result, we expected better uniformity in the lateral direction from the 3 in. target. The ratio of the erosion areas of the 2 in. and the 3 in. targets was found to be 1:1.8. Therefore, a rf power of 100 W was applied to the 2 in. target and 180 W to the 3 in. target.

YBCO films from both targets were deposited at room temperature on stationary 8 in. Si wafers. An Alpha Step 500 surface profiler was used to measure the thickness of the

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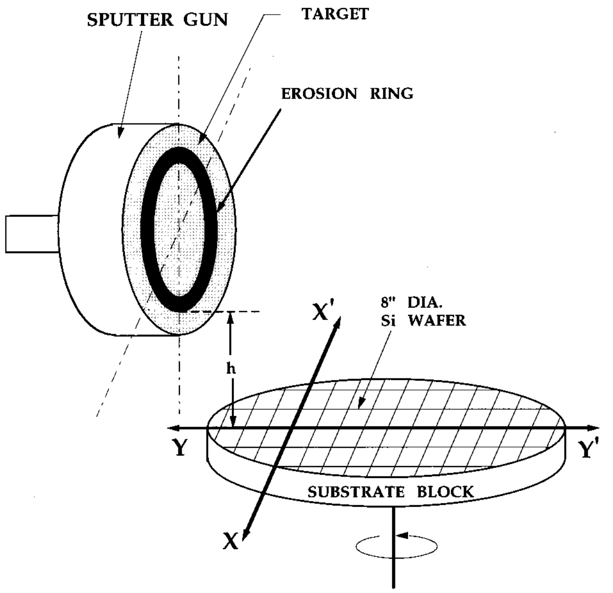

FIG. 1. The 90° off-axis sputtering geometry and the various reference axes on the substrates across which thickness, composition and T_c were measured.

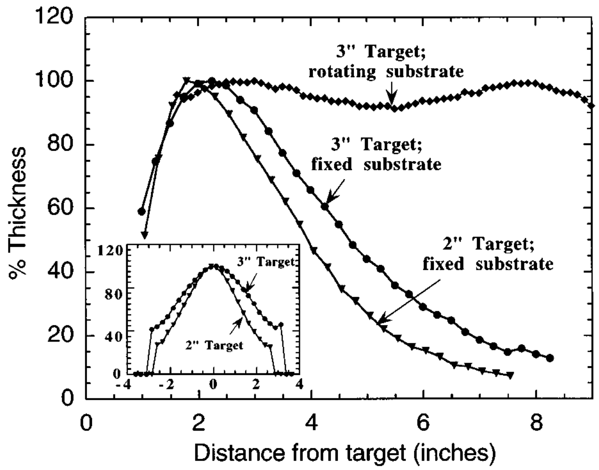

FIG. 2. Comparison of thickness profiles obtained from a 2 in. target and 3 in. target on a stationary substrate, along the $Y-Y'$ axis. The thickness profile obtained from the 3 in. target on a rotating substrate is superimposed for comparison. The inset shows a comparison of the thickness profiles along the $X-X'$ axis.

films at every 0.25 in. in both the $X-X'$ and $Y-Y'$ directions, as shown in Fig. 1. Thus, a two dimensional array consisting of about 900 measured thickness values was obtained which represented the complete map of the thickness distribution over an 8 in. diameter area. The thickness of the films ranged from 1000 Å to 6000 Å.

The thickness profiles measured along two orthogonal directions—the lateral $(X-X')$ axis and the longitudinal $(Y-Y')$ axis—are shown in Fig. 2. The distribution along the $X-X'$ axis is bell shaped and symmetric about the longitudinal axis because the ring shaped erosion zone is also symmetric about the same axis. Due to its larger erosion zone diameter, the thickness distribution obtained from the 3 in. target is more uniform than that obtained from the 2 in. target. Furthermore, the deposition rate obtained from the 3 in. target is about 33% higher than the rate from the 2 in. target.

The thickness of the film along the $Y-Y'$ direction increases across the substrate as the distance from the target increases until it reaches a peak value, beyond which the thickness decreases monotonically. This asymmetrical distribution leads us to believe that rotating the substrate would expose every part of the substrate to a deposition rate along the $Y-Y'$ axis which alternates between being high and low. Thus, the average over time should be a uniform thickness over a large area of the substrate. To obtain the maximum area with uniform thickness for a given target diameter, the optimum distance of the target from the center of substrate rotation needs to be identified. This was achieved using a computer simulation which uses the two dimensional thickness profile data obtained on a stationary substrate to produce the thickness distribution that would be obtained on a rotating substrate. The simulation uses an azimuthal averaging algorithm and iteratively solves for the optimum target distance. The program used a thickness uniformity requirement of ±5% to determine the area.

The thickness distribution predicted by the simulation was confirmed in an experiment where the substrate was continuously rotated during deposition at a speed of 12 rpm about the center of the wafer, as shown in Fig. 1. The thickness distribution obtained from the 3 in. target on a rotating substrate is shown, in comparison with that on a stationary substrate, in Fig. 2. A variation of less than ±5% in thickness is obtained over the entire 8-in.-diam. substrate, in contrast to the 1.3-in.-diam area obtained without substrate rotation.

Furthermore, the average deposition rates obtained from the 3-in.-diam. and 2-in.-diam. targets on rotating substrates were found to be 172 Å/h and 108 Å/h. This result clearly shows that the deposition rate as well as the thickness uniformity can be scaled up by increasing the target diameter in 90° off-axis sputtering.

The composition variation and stoichiometry of the films deposited on both stationary and rotating substrates were measured with Rutherford backscattering spectroscopy (RBS). On stationary substrates, a maximum composition variation of 3.2% along an 8 in. strip in the $Y-Y'$ direction was observed for films deposited from the 3 in. target. Along the $Y-Y'$ axis, the films deposited from both the 2 in. and 3 in. targets displayed an increase in Ba content and decrease in Cu content as the distance from the target was increased.

The films deposited from the 3 in. target onto a rotating substrate displayed a maximum variation in composition of less than 2.5% over an 8-in.-diam area, as shown in Fig. 3. The percentages mentioned here refer to the sum of Y, Ba, and Cu content as 100%. The maximum deviation from the target stoichiometry was observed to be +1.2% in Y, +1.5% in Ba, and −2.2% in Cu.

The variation in superconducting transition temperature and critical current density was measured as a function of substrate position with rotation. A 3-in.-diam. resistive substrate heater mounted on a rotating arm was used to grow crystalline YBCO thin films at high temperature. The radial distance of the heater from the center of rotation was in-

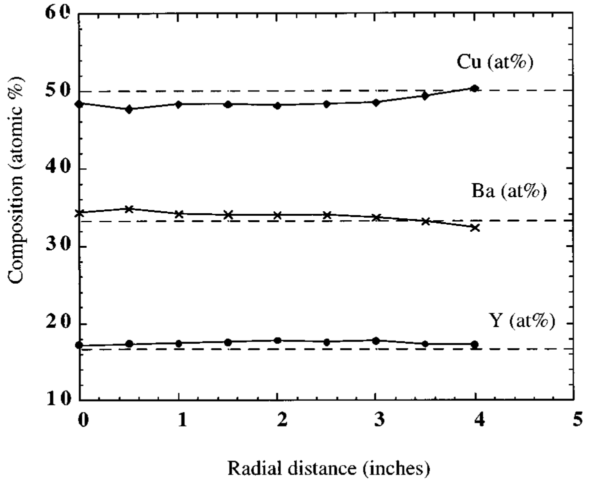

FIG. 3. The variation in composition obtained from the 3 in. target on a rotating substrate. The composition of the target, shown in broken lines, is superimposed for comparison.

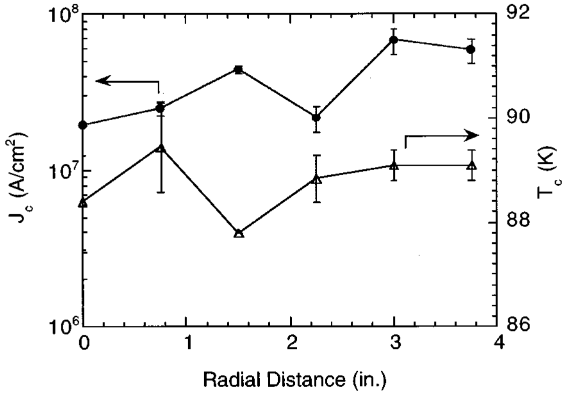

FIG. 4. Variation in T_c and J_c at 4.2 K of 4000-Å-thick YBCO films deposited from the 3 in. target as a function of the radial distance, r, from the center of rotation. The bars represent the spread in the data obtained from the measurement of two samples at each value of r.

creased for successive runs until the entire 8-in.-diam. area was studied. All the films for these studies were deposited on 0.25 in.×0.25 in. $LaAlO_3$ substrates at 735 °C. The transition temperature (T_c) of the films deposited from the 3-in.-diam. target was consistently high (>87.5 K) over a radial distance of 4 in. (i.e., 8-in.-diam. area), as shown in Fig. 4. We believe that the variation in the transition temperatures is a result of the variation in substrate temperature on the heater block and not a result of variation in composition.

The critical current density (J_c) of the films was measured by the dc magnetization method. The magnetization of the films was measured as a function of the applied field in the range of ±1 T in an Oxford Instruments MAGLAB vibrating sample magnetometer system. For films deposited from the 3 in. target over a 4 in. radial distance, the J_c near zero field at 4.2 K was consistently greater than 2×10^7 A/cm^2, as shown in Fig. 4(c). It is expected that a similar variation in T_c and J_c will be obtained along any radius of the 8-in.-diam. area because the variation of properties on rotating substrates should be symmetric about the center of rotation. Thus, the variation over a 4 in. radial distance is representative of the distribution over an 8-in.-diam. area.

The surface morphology of 1000-Å-thick films deposited on $LaAlO_3$ substrates, at different radial distances from the center of rotation was examined with an atomic force microscope (AFM). All the films were scanned over a 2 μm×2 μm area in the contact mode. Typical growth spirals were observed on the YBCO film indicating that the films grew via a screw dislocation mediated growth. The root mean square surface roughness of the film varied between 46 and 134 Å over a 4 in. radial distance.

We have demonstrated the deposition of high quality YBCO thin films by a 90° off-axis sputtering technique over very large areas (8 in. in diameter). Complete two dimensional mapping of the thickness distribution was obtained from a 2-in.- and a 3-in.-diam. target in a 90° off-axis sputtering geometry. As the diameter of the target is increased better uniformity can be obtained at a higher growth rate. This is due to more lateral spread of the flux and the increase in flux emitted from the larger erosion area of the target. The optimum distance of the target from the center of substrate rotation can be predicted by the computer simulation for any desired maximum variation in thickness. Hence, by decreasing the area of optimization, the uniformity in thickness, composition, and superconducting properties can be further improved. The results obtained here are technologically important because the same approach can also be applied for large area deposition of other oxide thin films.

We would like to thank Dr. Howle for his help on this project, Herb Kott at US Thin Film Products Inc. for providing the sputter guns for these experiments. Z. Xu and A. Guloy at the University of Houston for the magnetization measurements. This work was supported by the NSF grant No. DMR 9421947, the ONR grant No. N00014-95-1-0513, the NSF Young Investigator Award (CBE), and the David and Lucile Packard Fellowship (CBE).

[1] S. R. Foltyn, R. E. Muenchausen, R. C. Dye, X. D. Wu, L. Luo, D. W. Cooke, and R. C. Taber, Appl. Phys. Lett. **59**, 1374 (1991).
[2] C. S. Chern, J. S. Martens, Y. Q. Li, B. M. Gallois, P. Lu, and B. H. Kear, Supercond. Sci. Technol. **6**, 460 (1993).
[3] X. X. Xi, T. Venkatesan, Q. Li, X. D. Wu, A. Inam, C. C. Chang, R. Ramesh, D. M. Hwang, T. S. Ravi, A. Findikoglu, D. Hemmick, S. Etemad, J. A. Martinez, and E. Wilkens, IEEE Trans. Magn. **27**, 982 (1991).
[4] U. Krüger, R. Kutzner, and R. Wördenweber, IEEE Trans. Appl. Supercond. **3**, 1687 (1993).
[5] N. Newman, B. F. Cole, S. M. Garrison, K. Char, and R. C. Taber, IEEE Trans. Magn. **27**, 1276 (1991).
[6] C. B. Eom, J. Z. Sun, K. Yamamoto, A. F. Marshall, K. E. Luther, S. S. Laderman, and T. H. Geballe, Appl. Phys. Lett. **55**, 595 (1989).
[7] D. J. Lichtenwalner, C. N. Soble II, R. R. Woolcott, Jr., O. Auciello, and A. I. Kingon, J. Appl. Phys. **70**, 6952 (1991).
[8] D. W. Face, C. Wilker, Z.-Y. Shen, P. Pang, and R. J. Small, IEEE Trans. Appl. Supercond. **5**, 1581 (1995).
[9] D. W. Face, C. Wilker, Z.-Y. Shen, and P. S. W. Pang, presented at the Materials Research Society-Spring 1996 Meeting, MRS Abstracts, Q11.1, p. 308.
[10] H. Kinder, P. Berberich, W. Prusseit, R. Semerad, and B. Utz, presented at the International Workshop on Superconductivity June 1995, Program and Extended Abstracts, RII-5, p. 182.
[11] C. B. Eom, J. Z. Sun, S. K. Streiffer, A. F. Marshall, K. Yamamoto, B. M. Lairson, S. M. Anlage, J. C. Bravman, T. H. Geballe, S. S. Laderman, and R. C. Taber, Physica C **171**, 351 (1990).